

WHAT IS CLAIMED IS:

1. A magnetic recording medium comprising:  
a non-magnetic base film; and  
a magnetic recording layer comprising a binder resin and  
black magnetic acicular composite particles having an average  
particle diameter of 0.051 to 0.35  $\mu\text{m}$ , comprising  
magnetic acicular cobalt-coated iron oxide particles or  
magnetic acicular metal particles containing iron as a main  
component,  
a coating formed on surface of said magnetic acicular  
particles, comprising at least one organosilicon compound  
selected from the group consisting of:  
(1) organosilane compounds obtainable from alkoxy silane  
compounds, and  
(2) polysiloxanes or modified polysiloxanes, and  
a carbon black coat formed on said coating layer  
comprising said organosilicon compound, in an amount of from  
more than 10 to 40 parts by weight based on 100 parts by  
weight of said magnetic acicular particles.

2. A magnetic recording medium according to claim 1,  
wherein said magnetic acicular particles are particles having  
a coat which is formed on at least a part of the surface of  
said magnetic acicular particles and which comprises at least  
one compound selected from the group consisting of hydroxides

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of aluminum, oxides of aluminum, hydroxides of silicon and oxides of silicon in an amount of 0.01 to 20 % by weight, calculated as Al or  $\text{SiO}_2$ , based on the total weight of the magnetic acicular particles coated.

3. A magnetic recording medium according to claim 1, wherein said modified polysiloxanes are compounds selected from the group consisting of:

(A) polysiloxanes modified with at least one compound selected from the group consisting of polyethers, polyesters and epoxy compounds, and

(B) polysiloxanes whose molecular terminal is modified with at least one group selected from the group consisting of carboxylic acid groups, alcohol groups and a hydroxyl group.

4. A magnetic recording medium according to claim 1, wherein said alkoxy silane compound is represented by the general formula (I):

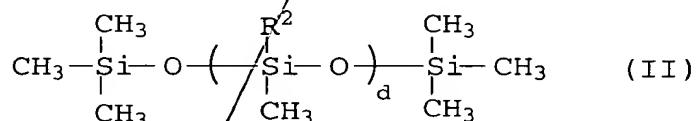


wherein  $\text{R}^1$  is  $\text{C}_6\text{H}_5-$ ,  $(\text{CH}_3)_2\text{CHCH}_2-$  or  $n\text{C}_b\text{H}_{2b+1}-$  (wherein  $b$  is an integer of 1 to 18);  $\text{X}$  is  $\text{CH}_3\text{O}-$  or  $\text{C}_2\text{H}_5\text{O}-$ ; and  $a$  is an integer of 0 to 3.

5. A magnetic recording medium according to claim 4, wherein said alkoxy silane compound is methyl triethoxysilane,

dimethyl diethoxysilane, phenyl triethoxysilane, diphenyl diethoxysilane, methyl trimethoxysilane, dimethyl dimethoxysilane, phenyl trimethoxysilane, diphenyl dimethoxysilane, isobutyl trimethoxysilane or decyl trimethoxysilane.

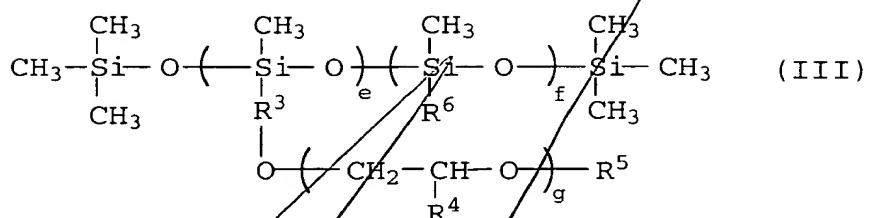
6. A magnetic recording medium according to claim 1, wherein said polysiloxanes are represented by the general formula (II):



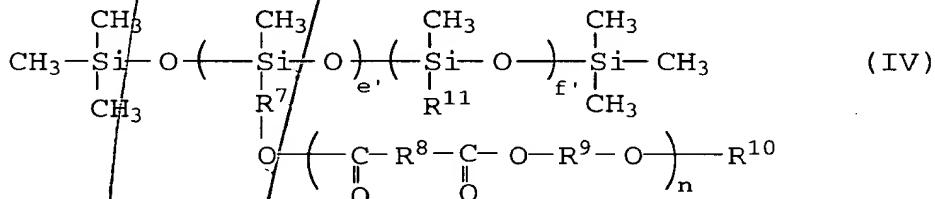
wherein  $\text{R}^2$  is  $\text{H-}$  or  $\text{CH}_3-$ , and  $d$  is an integer of 15 to 450.

7. A magnetic recording medium according to claim 6, wherein said polysiloxanes are compounds having methyl hydrogen siloxane units.

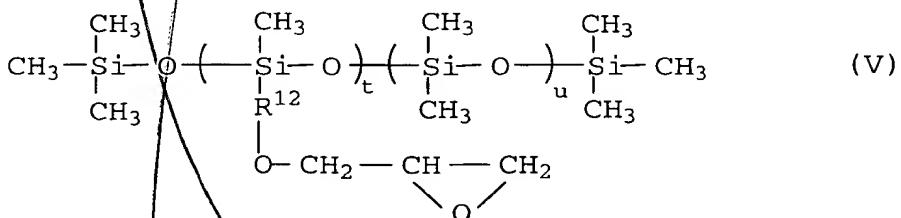
8. A magnetic recording medium according to claim 3, wherein said polysiloxanes modified with at least one compound selected from the group consisting of polyethers, polyesters and epoxy compounds are represented by the general formula (III), (IV) or (V):



wherein R<sup>3</sup> is  $-(\text{CH}_2-)_h-$ ; R<sup>4</sup> is  $-(\text{CH}_2-)_i\text{CH}_3$ ; R<sup>5</sup> is  $-\text{OH}$ ,  $-\text{COOH}$ ,  $-\text{CH}=\text{CH}_2$ ,  $-\text{C}(\text{CH}_3)=\text{CH}_2$  or  $-(\text{CH}_2-)_j\text{CH}_3$ ; R<sup>6</sup> is  $-(\text{CH}_2-)_k\text{CH}_3$ ; g and h are an integer of 1 to 15; i, j and k are an integer of 0 to 15; e is an integer of 1 to 50; and f is an integer of 1 to 300;

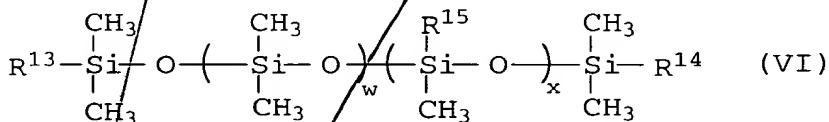


wherein R<sup>7</sup>, R<sup>8</sup> and R<sup>9</sup> are  $-(\text{CH}_2-)_q-$  and may be the same or different; R<sup>10</sup> is  $-\text{OH}$ ,  $-\text{COOH}$ ,  $-\text{CH}=\text{CH}_2$ ,  $-\text{C}(\text{CH}_3)=\text{CH}_2$  or  $-(\text{CH}_2-)_r\text{CH}_3$ ; R<sup>11</sup> is  $-(\text{CH}_2-)_s\text{CH}_3$ ; n and q are an integer of 1 to 15; r and s are an integer of 0 to 15; e' is an integer of 1 to 50; and f' is an integer of 1 to 300; or



wherein  $R^{12}$  is  $-(-CH_2-)_v-$ ;  $v$  is an integer of 1 to 15;  $t$  is an integer of 1 to 50; and  $u$  is an integer of 1 to 300.

9. A magnetic recording medium according to claim 3, wherein said polysiloxanes whose molecular terminal is modified with at least one group selected from the group consisting of carboxylic acid groups, alcohol groups and a hydroxyl group are represented by the general formula (VI):



wherein  $R^{13}$  and  $R^{14}$  are  $-OH$ ,  $R^{16}OH$  or  $R^{17}COOH$  and may be the same or different;  $R^{15}$  is  $-CH_3$  or  $-C_6H_5$ ;  $R^{16}$  and  $R^{17}$  are  $-(-CH_2-)_y-$ ;  $y$  is an integer of 1 to 15;  $w$  is an integer of 1 to 200; and  $x$  is an integer of 0 to 100.

10. A magnetic recording medium according to claim 1, wherein the amount of said coating organosilicon compounds is 0.02 to 5.0 % by weight, calculated as Si, based on the total weight of the organosilicon compounds and said magnetic acicular particles.

11. A magnetic recording medium according to claim 1,

wherein said carbon black coat is obtained by mixing carbon black fine particles having a particle size of 0.005 to 0.05  $\mu\text{m}$  with the magnetic acicular particles coated with at least one organosilicon compound while applying a shear force.

12. A magnetic recording medium according to claim 1, wherein the thickness of said carbon black coat is not more than 0.06  $\mu\text{m}$ .

13. A magnetic recording medium according to claim 1, wherein said black magnetic acicular composite particles have an absorption amount of myristic acid of 0.01 to 0.3  $\text{mg}/\text{m}^2$ .

14. A magnetic recording medium according to claim 1, wherein said black magnetic acicular composite particles have an aspect ratio (average major axis diameter/average minor axis diameter) of 2.0:1 to 20.0:1.

15. A magnetic recording medium according to claim 1, wherein said black magnetic acicular composite particles have a BET specific surface area value of 21 to 160  $\text{m}^2/\text{g}$ .

16. A magnetic recording medium according to claim 1, wherein said black magnetic acicular composite particles have a blackness ( $L^*$  value) of 15 to 22.

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17. A magnetic recording medium according to claim 1, wherein said black magnetic acicular composite particles have a volume resistivity of not more than  $1.0 \times 10^6 \Omega \cdot \text{cm}$ .

18. A magnetic recording medium according to claim 1, wherein said black magnetic acicular composite particles have a geometrical standard deviation of major axis diameter of 1.01 to 2.0.

19. A magnetic recording medium according to claim 1, which further comprises a gloss of coating film of 165 to 300 %, a surface roughness Ra of coating film of not more than 11.5 nm, a linear absorption of coating film of 1.75 to 10.0  $\mu\text{m}^{-1}$ , and a surface electrical resistivity of not more than  $1.0 \times 10^8 \Omega/\text{cm}^2$ .

20. A magnetic recording medium according to claim 1 or 2, which further comprises a non-magnetic undercoat layer disposed between said non-magnetic base film and said magnetic recording layer.

21. A magnetic recording medium according to claim 20, which further comprises a gloss of coating film of 165 to 300 %, a surface roughness Ra of coating film of not more than 11.0 nm, a linear absorption of coating film of 1.80 to 10.0  $\mu\text{m}^{-1}$ , and a surface electrical resistivity of not more than  $1.0 \times 10^8 \Omega/\text{cm}^2$ .

$\mu\text{m}^{-1}$ , and a surface electrical resistivity of not more than  $1.0 \times 10^8 \Omega/\text{cm}^2$ .

22. Black magnetic acicular composite particles for a magnetic recording medium, said black magnetic acicular composite particles having an average particle diameter of 0.051 to 0.35  $\mu\text{m}$ , comprising:

magnetic acicular cobalt-coated iron oxide particles or magnetic acicular metal particles containing iron as a main component,

a coating formed on surface of said magnetic acicular particles, comprising at least one organosilicon compound selected from the group consisting of:

(1) organosilane compounds obtainable from alkoxysilane compounds, and

(2) polysiloxanes or modified polysiloxanes, and

a carbon black coat formed on said coating layer comprising said organosilicon compound, in an amount of from more than 10 to 40 parts by weight based on 100 parts by weight of said magnetic acicular particles.

23. Black magnetic acicular composite particles according to claim 22, wherein said magnetic acicular particles are particles having a coat which is formed on at least a part of the surface of said magnetic acicular particles and which

comprises at least one compound selected from the group consisting of hydroxides of aluminum, oxides of aluminum, hydroxides of silicon and oxides of silicon in an amount of 0.01 to 20 % by weight, calculated as Al or SiO<sub>2</sub>, based on the total weight of the magnetic acicular particles coated.

24. Black magnetic acicular composite particles according to claim 22, wherein the thickness of said carbon black coat is not more than 0.06  $\mu\text{m}$ .

25. Black magnetic acicular composite particles according to claim 22, wherein said black magnetic acicular composite particles have an aspect ratio (average major axis diameter/average minor axis diameter) of 2.0:1 to 20.0:1, a BET specific surface area value of 21 to 160  $\text{m}^2/\text{g}$ , a blackness ( $L^*$  value) of 15 to 22, a volume resistivity of not more than  $1.0 \times 10^6 \Omega \cdot \text{cm}$ , and a geometrical standard deviation of major axis diameter of 1.01 to 2.0.

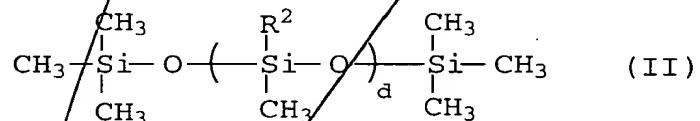
26. Black magnetic acicular composite particles according to claim 22, wherein said alkoxy silane compound is represented by the general formula (I):



wherein  $\text{R}^1$  is  $\text{C}_6\text{H}_5-$ ,  $(\text{CH}_3)_2\text{CHCH}_2-$  or  $n\text{-C}_b\text{H}_{2b+1}-$  (wherein  $b$  is an

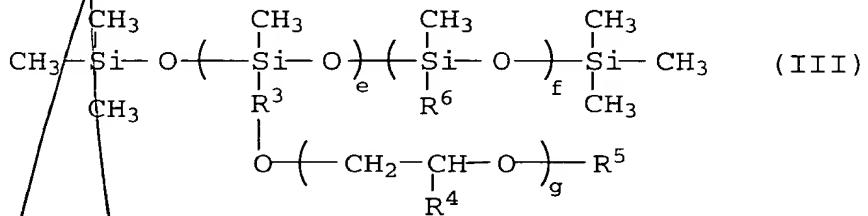
integer of 1 to 18); X is  $\text{CH}_3\text{O}^-$  or  $\text{C}_2\text{H}_5\text{O}^-$ ; and a is an integer of 0 to 3.

27. Black magnetic acicular composite particles according to claim 22, wherein said polysiloxanes are represented by the general formula (II):



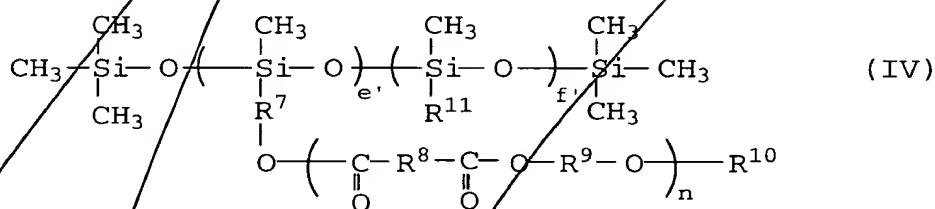
wherein R<sup>2</sup> is H- or CH<sub>3</sub>-, and d is an integer of 15 to 450.

28. Black magnetic acicular composite particles according to claim 22, wherein said polysiloxanes modified with at least one compound selected from the group consisting of polyethers, polyesters and epoxy compounds are represented by the general formula (III), (IV) or (V):



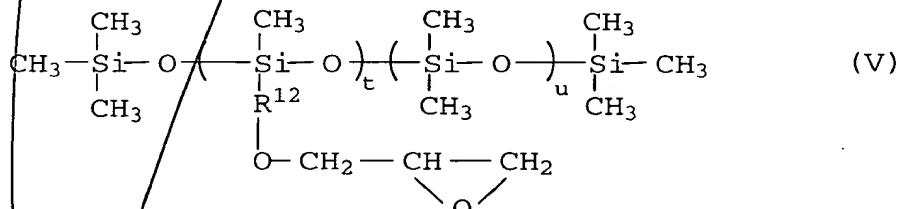
wherein  $R^3$  is  $\text{--}(-\text{CH}_2\text{--})_h\text{--}$ ;  $R^4$  is  $\text{--}(-\text{CH}_2\text{--})_i\text{--CH}_3$ ;  $R^5$  is  $\text{--OH}$ ,  $\text{--COOH}$ ,  $\text{--CH=CH}_2$ ,  $\text{--C(CH}_3\text{)}=\text{CH}_2$  or  $\text{--}(-\text{CH}_2\text{--})_j\text{--CH}_3$ ;  $R^6$  is  $\text{--}(-\text{CH}_2\text{--})_k\text{--CH}_3$ ;  $g$  and

h are an integer of 1 to 15; i, j and k are an integer of 0 to 15; e is an integer of 1 to 50; and f is an integer of 1 to 300;



wherein R<sup>7</sup>, R<sup>8</sup> and R<sup>9</sup> are  $-(\text{CH}_2-)_q-$  and may be the same or different; R<sup>10</sup> is -OH, -COOH, -CH=CH<sub>2</sub>, -C(CH<sub>3</sub>)=CH<sub>2</sub> or  $-(\text{CH}_2-)_r\text{CH}_3$ ; R<sup>11</sup> is  $-(\text{CH}_2-)_s\text{CH}_3$ ; n and q are an integer of 1 to 15; r and s are an integer of 0 to 15; e' is an integer of 1 to 50; and f' is an integer of 1 to 300; or

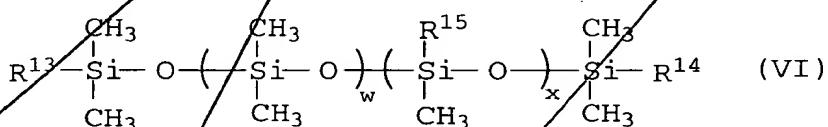
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wherein R<sup>12</sup> is  $-(\text{CH}_2-)_v-$ ; v is an integer of 1 to 15; t is an integer of 1 to 50; and u is an integer of 1 to 300.

29. Black magnetic acicular composite particles according to claim 22, wherein said polysiloxanes whose molecular terminal is modified with at least one group

selected from the group consisting of carboxylic acid groups, alcohol groups and a hydroxyl group are represented by the general formula (VI):



wherein  $\text{R}^{13}$  and  $\text{R}^{14}$  are  $-\text{OH}$ ,  $\text{R}^{16}\text{OH}$  or  $\text{R}^{17}\text{COOH}$  and may be the same or different;  $\text{R}^{15}$  is  $-\text{CH}_3$  or  $-\text{C}_6\text{H}_5$ ;  $\text{R}^{16}$  and  $\text{R}^{17}$  are  $-(-\text{CH}_2-)_y-$ ;  $y$  is an integer of 1 to 15;  $w$  is an integer of 1 to 200; and  $x$  is an integer of 0 to 100.

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30. Black magnetic acicular composite particles according to claim 22, wherein the amount of said coating organosilicon compounds is 0.02 to 5.0 % by weight, calculated as Si, based on the total weight of the organosilicon compounds and said magnetic acicular particles.

31. In a method of forming a magnetic recording medium comprising a non-magnetic base film, and a magnetic recording layer comprising a binder resin and magnetic particles, the improvement comprising using as magnetic particles black magnetic acicular composite particles having an average particle diameter of 0.051 to 0.35  $\mu\text{m}$ , comprising magnetic acicular cobalt-coated iron oxide particles or

magnetic acicular metal particles containing iron as a main component,

a coating formed on surface of said magnetic acicular particles, comprising at least one organosilicon compound selected from the group consisting of:

(1) organosilane compounds obtainable from alkoxysilane compounds, and

(2) polysiloxanes or modified polysiloxanes, and

a carbon black coat formed on said coating layer comprising said organosilicon compound, in an amount of from more than 10 to 40 parts by weight based on 100 parts by weight of said magnetic acicular particles.

32. The method according to claim 31, wherein said magnetic acicular particles are particles having a coat which is formed on at least a part of the surface of said magnetic acicular particles and which comprises at least one compound selected from the group consisting of hydroxides of aluminum, oxides of aluminum, hydroxides of silicon and oxides of silicon in an amount of 0.01 to 20 % by weight, calculated as Al or  $\text{SiO}_2$ , based on the total weight of the magnetic acicular particles coated.

33. A magnetic recording medium comprising:

a non-magnetic base film;

a non-magnetic undercoat layer formed on said non-magnetic base film; and

a magnetic recording layer comprising a binder resin and black magnetic acicular composite particles having an average particle diameter of 0.051 to 0.35  $\mu\text{m}$ , comprising

magnetic acicular cobalt-coated iron oxide particles or magnetic acicular metal particles containing iron as a main component,

a coating formed on surface of said magnetic acicular particles, comprising at least one organosilicon compound selected from the group consisting of:

(1) organosilane compounds obtainable from alkoxy silane compounds, and

(2) polysiloxanes or modified polysiloxanes, and

a carbon black coat formed on said coating layer comprising said organosilicon compound, in an amount of 0.5 to 10 parts by weight based on 100 parts by weight of said magnetic acicular particles.

34. A magnetic recording medium according to claim 33, wherein said magnetic acicular particles are particles having a coat which is formed on at least a part of the surface of said magnetic acicular particles and which comprises at least one compound selected from the group consisting of hydroxides of aluminum, oxides of aluminum, hydroxides of silicon and oxides of silicon in an amount of 0.01 to 20 % by weight,

calculated as Al or SiO<sub>2</sub>, based on the total weight of the magnetic acicular particles coated.

35. A magnetic recording medium according to claim 33, wherein said modified polysiloxanes are compounds selected from the group consisting of:

(A) polysiloxanes modified with at least one compound selected from the group consisting of polyethers, polyesters and epoxy compounds, and

(B) polysiloxanes whose molecular terminal is modified with at least one group selected from the group consisting of carboxylic acid groups, alcohol groups and a hydroxyl group.

36. A magnetic recording medium according to claim 33, wherein said alkoxy silane compound is represented by the general formula (I):



wherein R<sup>1</sup> is C<sub>6</sub>H<sub>5</sub>- or (CH<sub>3</sub>)<sub>2</sub>CHCH<sub>2</sub>- or n-C<sub>b</sub>H<sub>2b+1</sub>- (wherein b is an integer of 1 to 18); X is CH<sub>3</sub>O- or C<sub>2</sub>H<sub>5</sub>O-; and a is an integer of 0 to 3.